



Europäisches Patentamt
European Patent Office
Office européen des brevets



(11) **EP 0 724 069 A2**

(12)

EUROPEAN PATENT APPLICATION

(43) Date of publication:
31.07.1996 Bulletin 1996/31

(51) Int. Cl.⁶: **F01N 3/28**

(21) Application number: 96250086.4

(22) Date of filing: 10.05.1993

(84) Designated Contracting States:
AT BE CH DE DK ES FR GB GR IE IT LI LU MC NL
PT SE

(30) Priority: 19.05.1992 US 885456

(62) Application number of the earlier application in
accordance with Art. 76 EPC: 93250131.5

(71) Applicant: W.R. Grace & Co.-Conn.
New York, New York 10036 (US)

(72) Inventor: Retallick, William B.
West Chester, PA 19382 (US)

(74) Representative: UEXKÜLL & STOLBERG
Patentanwälte
Beselerstrasse 4
22607 Hamburg (DE)

Remarks:

This application was filed on 15 - 04 - 1996 as a
divisional application to the application mentioned
under INID code 62.

(54) Method of making a catalytic reactor

(57) A catalytic reactor serves as the ignition stage for a multi-stage combustor. In one embodiment, the catalytic reactor includes a stack of corrugated metal strips, each strip having a catalyst coating on exactly one side, wherein a coated side of one strip faces an uncoated side of the adjacent strip. Thus, the reactor defines channels each having two walls, exactly one of which bears a catalyst coating. Another embodiment takes the form of a spiral formed by winding two strips, each strip having a catalyst coating on one side. The

coated side of one strip faces the uncoated side of the other. Thus, the spiral includes channels having a catalyst coating on only one wall. In still another embodiment, a single strip has bands of coated and uncoated regions on both sides. At any given point on the strip, one side of the strip has a catalyst coating and the other side has no coating. One forms the catalytic reactor by folding the strip back and forth upon itself, in a zigzag pattern, and enclosing the folded strip within a canister.

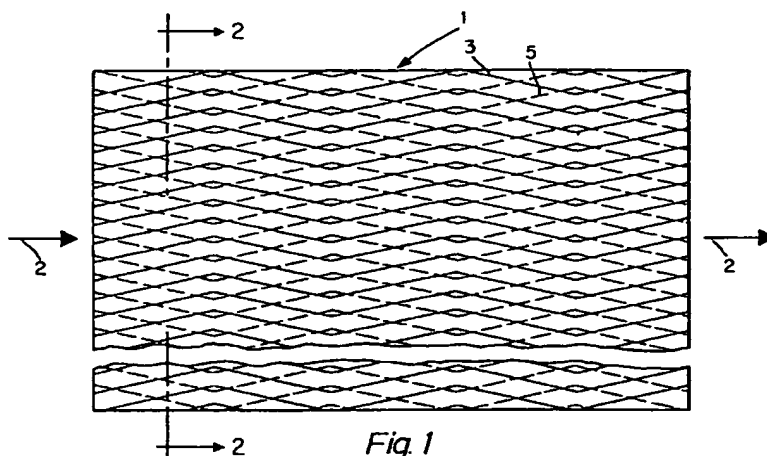


Fig 1

EP 0 724 069 A2

Description

BACKGROUND OF THE INVENTION

The present invention provides a combustion apparatus for use in a high-temperature environment, such as in a gas turbine, or elsewhere. More particularly, the invention includes a catalytic combustor which one can use as the initial stage in a multi-stage high-temperature combustion apparatus.

The present invention represents an improvement over the product and method described in copending U.S. patent application Serial No. 07/408,521, filed September 18, 1989, entitled "Combustion Apparatus for High-temperature Environment". This specification hereby incorporates the disclosure of the latter application by reference.

When a fuel-air mixture flows through the channels of a catalytic reactor, the temperature of the mixture increases smoothly as the combustion proceeds. But the temperature of the catalyst-coated wall of the channel does not increase smoothly. Instead, this temperature increases rapidly near the entrance to the channel and approaches the adiabatic combustion temperature. Then it remains close to the adiabatic combustion temperature over the length of the channel.

If one makes the channel too short to achieve complete combustion, the exiting gas will have a temperature below the adiabatic combustion temperature. Even so, the catalyst-coated wall will have a temperature close to adiabatic, over all but the front end of the channel. Thus, even with a shortened catalytic combustion channel, the catalyst-coated walls still reach the adiabatic temperature, thereby deactivating the catalyst.

The above-cited patent application discloses one solution to the above problem. The latter application shows a catalytic reactor made of a single strip of metal coated with catalyst on one side only. One embodiment includes a strip folded back and forth upon itself, in a zigzag pattern, to form a catalytic reactor. In another embodiment, either one or two strips become wound into a spiral. In either case, the catalytic reactor defines channels for gas flow in which a catalyst coats either all or none of each channel.

The present invention provides a modified structure which more evenly distributes the coated and uncoated portions of the metal strip. The invention also includes a method for making a catalytic reactor. The present invention prevents the walls of the combustor from reaching the adiabatic combustion temperature, as compared with catalytic combustors of the prior art.

SUMMARY OF THE INVENTION

The present invention includes a structure for the ignition stage of a multiple-stage combustion apparatus. One forms this ignition stage from one or more strips of metal, preferably having the thickness of a foil. The foil contains corrugations, the corrugations preferably

defining a herringbone or "chevron" pattern. Multiple layers of corrugated foil form a structure called a honeycomb. The corrugations in adjacent layers cross over each other and prevent the layers from nesting together. The corrugations also form channels for gas flow.

An important feature of the present invention comprises coating just one wall of each channel with catalyst. The other wall remains uncoated. At any point on a wall of a channel, both sides of the foil can transfer heat to the gas stream. But only the coated side can combust fuel and generate heat. Therefore the temperature of the foil will settle at a temperature below the adiabatic combustion temperature, and the catalyst does not become deactivated.

One embodiment of the invention includes two strips of foil, both coated with catalyst on only one side. One winds these strips together to form a continuous spiral channel. Only one wall of the channel has a catalyst coating.

Another embodiment of the invention includes a plurality of separate strips of metal. One stacks the strips to define channels that contain a catalyst coating on only one wall of each channel.

Still another embodiment includes a reactor made from a single strip of metal. The strip contains bands of catalyst coating on both sides of the strip. The end of a band on one side marks the beginning of a band on the other side. In other words, at any given point on the strip, the strip has a catalyst coating on one side but not the other. One folds the strip back and forth upon itself, in a zigzag pattern, to form the catalytic reactor. The present invention includes various ways of varying the arrangement of the bands and/or the fold lines. All such variations include a strip having a catalyst coating distributed along the strip, to prevent the strip from overheating.

The present invention therefore has the principal object of providing a catalytic combustion apparatus for use in a high-temperature environment.

The invention has the further object of providing a method for making a catalytic combustion apparatus for use in a high-temperature environment.

The invention has the further object of providing a method for making a catalytic ignition stage which does not become so hot that its metal catalyst becomes deactivated.

The invention has the further object of extending the useful life of a catalytic reactor.

The invention has the further object of making it more practical to use a catalytic reactor in a high-temperature environment.

Those skilled in the art will recognize other objects and advantages of the present invention, from a reading of the following brief description of the drawings, the detailed description of the invention, and the appended claims.

BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 shows a partially fragmentary plan view of the top two layers of a stack of metal strips, each strip having corrugations forming a herringbone pattern, the stack of strips forming the ignition stage of a combustion apparatus made according to the present invention.

Figure 2 provides a cross-sectional view taken along the line 2-2 of Figure 1, and shows the channels formed by the layers of metal strips.

Figures 3a and 3b show plan views of both sides of a single metal strip, used to make a catalytic reactor according to another embodiment of the present invention. The strip contains bands of coated and uncoated regions, and at every point along the strip, either one side or the other (but not both) has a catalyst coating.

Figure 4 provides a fragmentary end view of a catalytic reactor made according to the present invention, wherein one has folded a metal strip back and forth upon itself to define a generally circular cross-section.

Figure 5 provides a fragmentary end view of a single folded metal strip, wherein one has folded the strip along the middle of a band of catalyst coating, and wherein the interval between folds equals the length of one of the bands.

Figure 6 provides a view similar to that of Figure 5, but wherein one has folded the strip along the edge of a band.

Figure 7 provides a view similar to that of Figure 5, but wherein one has folded the strip at a point between the middle of a band and the edge of a band.

Figure 8 provides a view similar to that of Figure 5, wherein one has folded the strip along the edge of a band, and where the interval between folds equals twice the length of one of the bands.

Figure 9 provides a view similar to that of Figure 5, but wherein the interval between folds equals one-half the length of one of the bands.

Figure 10 provides a view similar to that of Figure 5, but wherein the bands of catalyst coating have varying lengths.

Figure 11 shows a fragmentary end view of a catalytic reactor made according to an embodiment of the present invention which includes two strips of metal wound into a spiral.

Figure 12 provides a graph showing temperature profiles of the metal strip and of the gas stream in a catalytic combustor.

DETAILED DESCRIPTION OF THE INVENTION

The present invention comprises a multiple-stage combustion apparatus for use in a high-temperature environment, and a method for making the apparatus. One designates the first stage as the ignition stage. The ignition stage employs a combustion catalyst, such as a metal from the platinum group, which ignites the incoming fuel-air mixture. This ignition stage must burn the fuel-air mixture only partly, so that the temperature of

the gas leaving the first stage of the combustion apparatus does not exceed about 1500°F. This specification uses the term "ignition stage" synonymously with "catalytic combustor" or "catalytic reactor".

Copending application Serial No. 07/408,521, cited above, discloses a construction that ensures that the first stage only partly burns the fuel-air mixture. In the cited application, alternate channels in the honeycomb combustor bear a catalyst coating. The channels containing the coating have the coating on both walls of the channel. The channels having no coating bear no catalyst on either wall. The uncombusted fuel-air mixture flowing through the uncoated channels cools the walls of the coated channels.

The major difference between the present invention and the combustor shown in the cited patent application resides in the manner of coating the channels.

Figure 1 shows catalytic ignition stage 1 formed of metal having herringbone, or "chevron", corrugations. Although this figure resembles Figure 1 of the above-cited application, it represents a different structure, insofar as Figure 1 of the present invention indicates a stack of corrugated strips, and not a single strip folded back and forth upon itself. Preferably, one makes the strips from a foil, but the invention can work with strips of considerably greater thicknesses. One first corrugates each strip and then coats the strip with catalyst on only one side. One stacks these strips in layers to form the catalytic ignition stage of the present invention.

Figure 1 shows the top layer in full and the next layer in phantom. Reference numeral 3 indicates the corrugations in the top layer, and reference numeral 5 indicates the corrugations in the next preceding layer. Because the corrugations in adjacent layers have a generally out-of-phase relationship, the layers do not nest together. Thus, the corrugations provide means for maintaining at least a portion of the strip in spaced apart relation relative to an adjacent strip. Arrows 2 indicate the general direction of gas flow.

Figure 2 provides a cross-sectional view taken along the line 2-2 of Figure 1, showing the channels formed by the stack of strips. Figure 2 shows clearly that each strip has a catalyst coating 7 on only one side. The folded strip defines channels 9. Each channel comprises two walls, one wall bearing a catalyst coating and the other wall remaining uncoated. Thus, in the embodiment of Figures 1 and 2, half of each channel bears a catalyst coating, and the other half remains uncoated. In the view of Figure 2, gas flows in a direction perpendicular to the paper. Thus, about half of the gas encounters a coated wall and the remainder encounters an uncoated wall.

Note also that the combustion gas flows through zigzag channels defined by the herringbone corrugations, as shown in Figure 1. Thus, the gas almost never flows exactly parallel to arrows 2; the arrows show only the general direction of gas flow.

The zigzag path has the advantage of providing better heat and mass transfer. By definition, the length

of a zigzag path connecting two points exceeds that of a straight path connecting those points. Thus, with the zigzag path, the gas has more opportunity to contact the reactive walls of the channels, and more opportunity to transfer heat. Thus, one can achieve the same ignition effect with a shorter length of catalyst. The zigzag path can thus reduce the amount of catalyst required.

Note also that Figure 2 provides a picture of the strips only along the particular line 2-2 of Figure 1. If one had taken the cross-section at another point, the corrugations would have a different relative "phase". Due to the herringbone configuration, the adjacent layers will not nest together.

Note also that the combustion gas, while traveling in the general direction indicated by arrows 2, can still traverse the width of the reactor, in a direction generally perpendicular to the direction of gas flow, at most (but not necessarily all) locations. This transverse flow of gas also has advantages. At the expense of some pressure drop, it also provides a higher rate of mass transfer, which further enhances the efficiency of the ignition stage.

In another embodiment of the present invention, one forms the catalytic ignition stage from a single metal strip. In this embodiment, the strip contains bands of catalyst coating on both sides of the strip. Figures 3a and 3b illustrate such a strip. Figure 3a represents one side of the strip and Figure 3b represents the opposite side. Thus, at any point along the strip, one side of the strip bears a catalyst coating, while the other side remains bare. In Figures 3a and 3b, the coated and uncoated bands have the same lengths, but one can also vary the lengths of the bands. The term "length", as used with respect to the bands, refers to the dimension of the band as measured along the length of the strip.

One uses a strip prepared as indicated in Figures 3a and 3b to make the catalytic ignition stage shown in Figure 4. Specifically, one folds the strip back and forth upon itself in a zigzag manner. Figure 4 shows a strip folded back and forth to define a generally circular cross-section, and encased within canister 13. Figure 4 shows only some of the layers, but does not explicitly show the catalyst coating. One can form many other shapes by folding the strip back and forth upon itself, in the manner shown in U.S. Patent Nos. 4,576,800 and 4,838,067. This specification incorporates by reference the disclosures of the latter patents.

By varying the lengths of the coated and uncoated bands in Figures 3a and 3b, one can make final products having very different patterns of catalyst coating. Figures 5-10 provide examples. These figures show fragmentary cross-sectional views of several folds of a strip, similar to the view of Figure 2. However, note that Figures 5-10 differ from Figure 2 in that each strip in Figures 5-10 constitutes a single piece, folded back and forth upon itself, while the structure of Figure 2 comprises a plurality of separate stacked strips.

The two major parameters which one must consider in making a catalytic ignition stage from the strip of

Figure 3 include the placement of the fold lines and the distance between fold lines. Figures 5-7 illustrate three cases in which the interval between folds equals one band width (assuming that the bands have equal lengths). Thus, in Figure 5, one has placed the fold lines in the middle of each band. In Figure 6, one has placed the fold lines at the edges of the bands. In Figure 7, one locates the fold line at a point intermediate the middle and the edge of the band.

Figure 8 illustrates a case in which the distance between folds equals two band widths. One has located the fold line at the edge of the band.

Figure 9 illustrates a case in which the distance between folds equals one-half the width of the band. In this case, the fold lines appear at the edges and/or the middle of the bands; if fold lines appear at edges, other fold lines must appear at the middle.

Figure 10 illustrates a case in which the bands have variable lengths.

The reader will easily recognize that the cases described above do not exhaust all possibilities. One can construct many other arrangements by varying the band width and/or the position of the fold lines and/or the interval between folds. Thus, some channels may have catalyst coating on one of two walls. Other channels may have catalyst coating on parts of both walls. Some channels may have catalyst coating on both of two walls or neither of the two walls. In particular, one can make the structure of Figure 4 wherein every channel includes one fully coated wall and one fully bare wall, by varying the widths of the bands at the appropriate places, in a manner analogous to the method of locating perforations in a strip in U.S. Patent No. 4,576,800. Or one can build Figure 4 wherein varying portions of each layer of the folded strip contain catalyst. The present invention includes all such possibilities. All of the possible arrangements have one feature in common, namely that every part of the strip contains a catalyst coating on exactly one side.

Figure 11 shows another embodiment of the present invention. This embodiment comprises two strips 41 and 42 wound into a spiral. Both strips have a catalyst coating 43 on only one side. Before winding the strips to form the spiral of Figure 11, one must first arrange the strips so that the coated sides do not face each other. In other words, the coated side of one strip faces the uncoated side of the other strip, and vice versa. Then, when wound, the strips define channels having walls, only one of which bears a catalyst coating.

Alternatively, one can form strips 41 and 42 from a single strip by coating the strip on one side from the end of the strip to its midpoint, and by coating the other half of the strip from the midpoint to the other end, on the other side of the strip. Then, one folds the strip upon itself about a point near its midpoint, and winds the strip into a spiral. One obtains a structure almost identical to that obtained from separate strips, except that in this alternative embodiment a piece of metal joins the two strips at their ends. Thus, one should consider this alter-

native a direct equivalent of the embodiment discussed earlier.

In another embodiment, instead of using strips fully coated on one side, one or both of the strips have bands of catalyst coating, formed in the same manner described with respect to Figures 3a and 3b. In the most general case, the bands need not have uniform lengths. One can form the bands with random lengths. Thus, in this embodiment, one can achieve a random distribution of catalyst throughout the spiral.

As shown above, the present invention includes embodiments wherein exactly one wall of every channel has a catalyst coating, while the other wall remains uncoated. In these embodiments, at any point on a wall of a channel, both sides of the metal foil can transfer heat to the gas stream, but only the coated side can combust fuel and generate heat. Therefore the temperature of the foil will not rise to the adiabatic combustion temperature at only a short distance from the inlet. If the channels have sufficient length, combustion reaches completion, and the foil near the output end of the ignition stage will reach adiabatic temperature.

Figure 12 illustrates the relationship between temperature and distance along a channel for the catalytic ignition stage of the present invention as well as for those of the prior art. Curve 61 represents the temperature of the metal strip (foil) in a reactor wherein all of the walls bear a catalyst coating. As shown, the temperature reaches the adiabatic temperature at a short distance from the inlet. Curve 62 traces the temperature of the gas stream in this same reactor. This temperature rises smoothly and reaches adiabatic temperature further downstream. Curve 63 traces the temperature of the metal strip in a reactor of the present invention. The temperature rises more slowly, but if the channels have sufficient length, the foil will reach adiabatic temperature. Curve 64 traces the temperature of the gas stream in a reactor of the present invention. Here also, the gas reaches adiabatic temperature if the channels have sufficient length. In a reactor of the present invention, one makes the channels short enough to prevent attainment of adiabatic temperature, either on the metal strip or in the gas stream. To accomplish this objective, one cuts off the reactor of the present invention at the position indicated generally by line A-A. Combustion in the gas stream will not reach completion, and neither the foil nor the gas stream have reached adiabatic temperature.

U.S. Patent No. 4,711,009, also incorporated by reference into this specification, describes a continuous process for corrugating a metal strip, coating the strip with catalyst, and folding the strip upon itself to form a honeycomb. In the process of the latter patent, one coats the strip with catalyst on both sides, as shown in Figure 8 of that patent. In practicing the present invention, one must modify the process of latter patent to apply the catalyst coating only in the desired locations. For the embodiment of Figures 1 and 2, which includes a stack of individual corrugated strips, or the embodiment of Figure 11, which includes a spiral formed of two

such strips, one simply applies the coating to only one side of each strip, by spraying the strip on only one side, or by applying the coating with a roller contacting one side of the strip. One wets the roller with the washcoat or with a solution of the catalyst metal.

For the embodiments which use a strip of the type shown in Figures 3a and 3b, one needs to modify the process of U.S. Patent No. 4,711,009 in a different way. One could spray a washcoat and/or catalyst on either side of the strip by alternately activating nozzles disposed on either side of the strip. Alternatively, one could apply the washcoat and/or catalyst from only one side of the strip, and could either pass the strip twice through the line, or pass the strip through two similar processing areas, using equipment programmed to apply a coating to the different sides of the strip in the proper locations.

Note that, for clarity of illustration, Figures 1 and 2 do not show the canister that encloses the stack of strips, and Figure 11 also does not show a canister. In practice, one would use such a canister. Figure 4 explicitly shows a canister.

While the specification describes the invention with respect to certain embodiments and examples, the reader skilled in the art will recognize many possible variations. One should consider such variations within the spirit and scope of the following claims.

Claims

1. A method of making a catalytic reactor (1), the method being characterized by the steps of:

- a) coating a plurality of distinct strips of metal with a catalyst (7), each strip being coated on only one side, each strip having means (3, 5) for maintaining at least a portion of the strips in spaced apart relation when the strips are stacked, and
- b) stacking the strips upon each other, in a manner such that the catalyst coatings (7) of adjacent strips in the stack do not face each other, the resulting stack comprising the reactor (1), wherein the reactor (1) includes a plurality of channels (9), each channel being defined by a pair of walls, wherein exactly one of the walls of said pair has a catalyst coating (7).

2. A method of making a catalytic reactor, the method being characterized by the steps of:

- a) coating a pair of strips (41, 42) of metal with a catalyst (43), each strip being coated on only one side, each strip having means for maintaining at least a portion of the strips in spaced apart relation when the strips are laid over each other, and

- b) laying the strips against each other, such that a coated side of one strip faces an uncoated side of the other strip, and
- c) winding the strips (41, 42), as arranged in step (b), into a spiral, the spiral comprising the reactor, wherein the reactor includes a plurality of channels, each channel being defined by a pair of walls, wherein exactly one of the walls of said pair has a catalyst coating (43).
3. A method of making a catalytic reactor, the method being characterized by the steps of:
- a) coating alternate sides of a strip of metal with a catalyst (7), the strip having means (3, 5) for maintaining at least a portion of the strip in spaced apart relation from another portion of the strip when said portions are arranged adjacent to each other, the coating being performed such that at any point on the strip, one side of the strip is coated with catalyst and the other side is not coated, and
- b) folding the strip back and forth upon itself in a zigzag pattern, to produce the reactor.
4. The method of Claim 3, characterized in that it further comprises the step of enclosing the folded strip within a canister (13).
5. The method of Claims 3 or 4, characterized in that the coating is applied to the strip in bands of coated and uncoated regions, the bands extending across the entire width of the strip, and wherein the bands have equal lengths.
6. The method of Claim 5, characterized in that the folding is done along lines coincident with the edges of the bands.
7. The method of Claim 5, characterized in that the folding is done along lines which are not at the edges of the bands.
8. The method of Claim 5, characterized in that the distance between folds is in the range of one-half to two times the length of a band.
9. The method of Claim 3, characterized in that the coating is applied to the strip in bands of coated and uncoated regions, the bands extending across the entire width of the strip, and wherein the bands have unequal lengths.
10. A method of making a catalytic reactor, the method being characterized by the steps of:
- a) providing a pair of strips (41, 42) of metal,
- b) coating at least one of said strips in a manner such that at any point on the strip, one side

of the strip is coated with catalyst (43) and the other side is not coated, each strip having means for maintaining at least a portion of the strips in spaced apart relation when the strips are laid over each other,

c) laying the strips against each other, and

d) winding the strips, as arranged in step (c), into a spiral, the spiral comprising the reactor.

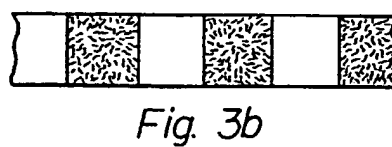
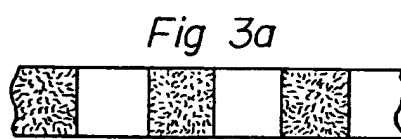
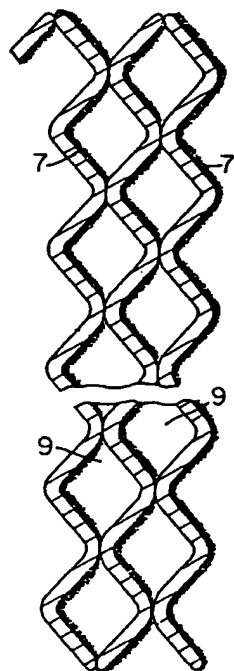
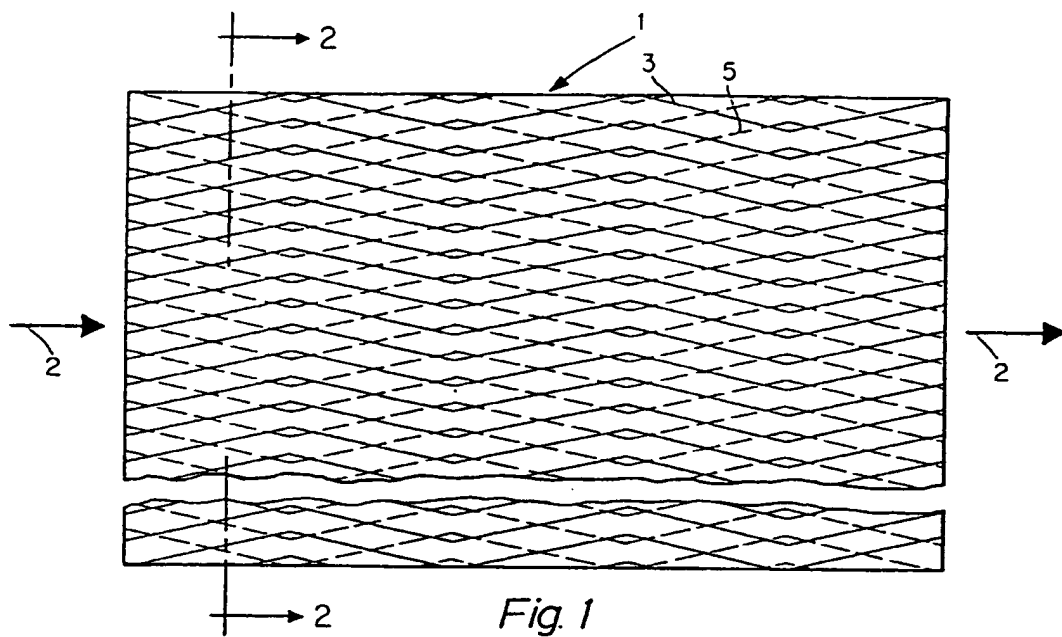


Fig. 4

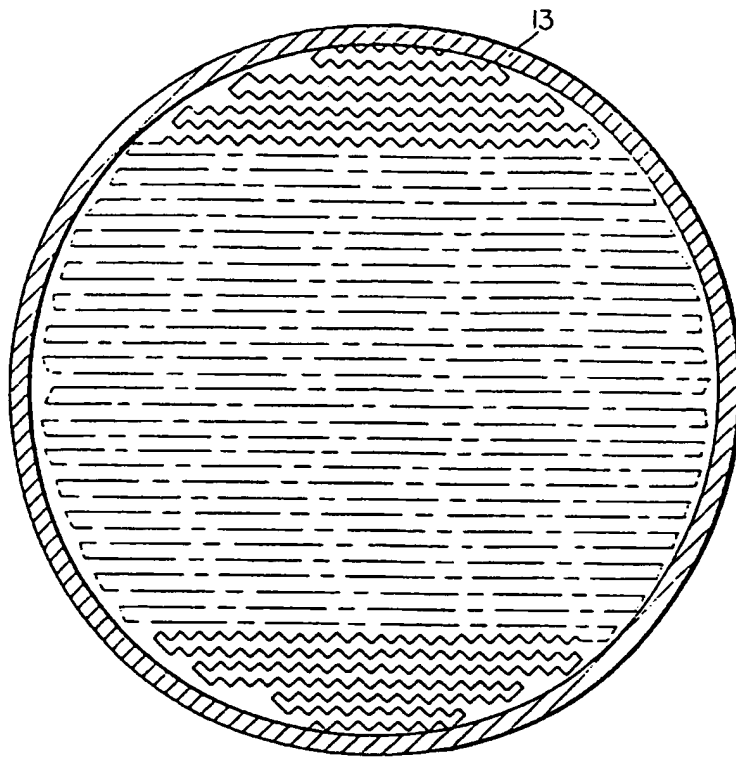


Fig. 5

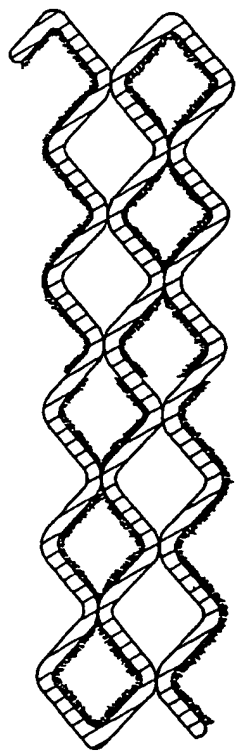


Fig. 6

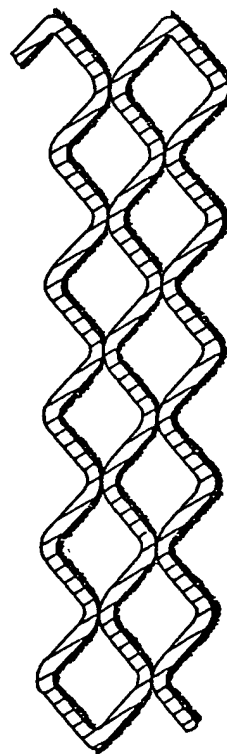


Fig. 7

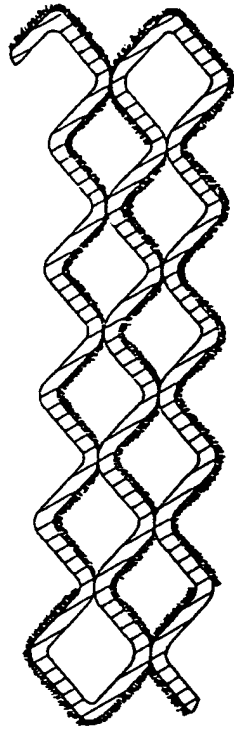


Fig. 8

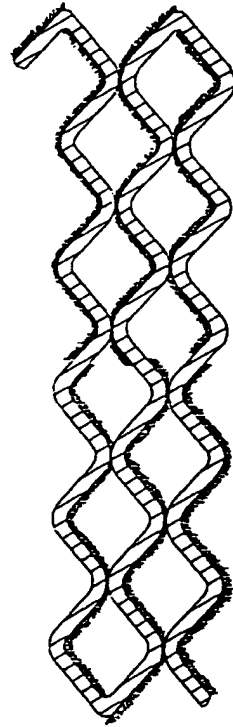


Fig. 9

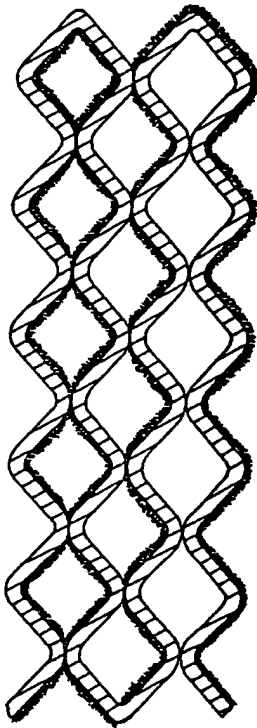


Fig. 10

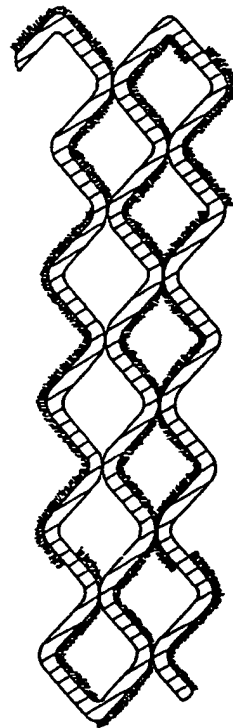


Fig. 11

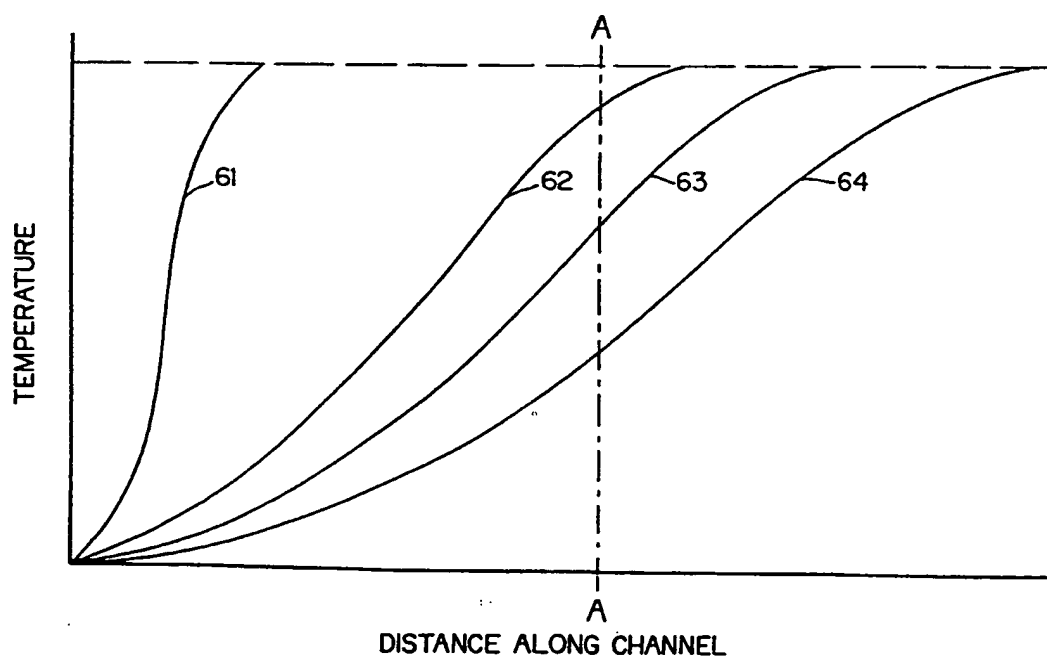
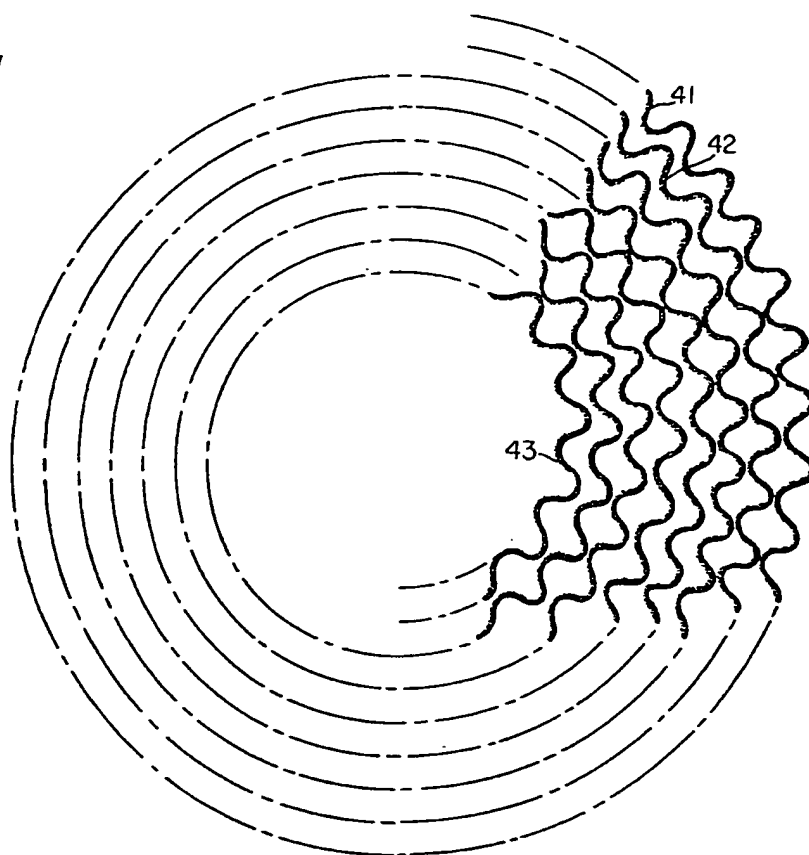


Fig. 12

PUB-NO: EP000724069A2

DOCUMENT-IDENTIFIER: EP 724069 A2

TITLE: Method of making a catalytic reactor

PUBN-DATE: July 31, 1996

INVENTOR-INFORMATION:

NAME	COUNTRY
RETALLICK, WILLIAM B	US

ASSIGNEE-INFORMATION:

NAME	COUNTRY
GRACE W R & CO	US

APPL-NO: EP96250086

APPL-DATE: May 10, 1993

PRIORITY-DATA: EP93250131A (May 10, 1993) , US88545692A (May 19, 1992)

INT-CL (IPC): F01N003/28

EUR-CL (EPC): F01N003/28 ; B01J035/04

ABSTRACT:

CHG DATE=19990617 STATUS=O> A catalytic reactor serves as the ignition stage for a multi-stage combustor. In one embodiment, the catalytic reactor includes a stack of corrugated metal strips, each strip having a catalyst coating on exactly one side, wherein a coated side of one strip faces an uncoated side of the adjacent strip. Thus, the reactor defines channels each having two walls, exactly one of which bears a catalyst coating. Another embodiment takes the form of a spiral formed by winding two strips, each strip having a catalyst

coating on one side. The coated side of one strip faces the uncoated side of the other. Thus, the spiral includes channels having a catalyst coating on only one wall. In still another embodiment, a single strip has bands of coated and uncoated regions on both sides. At any given point on the strip, one side of the strip has a catalyst coating and the other side has no coating. One forms the catalytic reactor by folding the strip back and forth upon itself, in a zigzag pattern, and enclosing the folded strip within a canister. <IMAGE>